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(54) GLASS LAMINATES

(71) We, IMPERIAL CHEMICAL INDUSTRIES LIMITED, of Imperial Chemical House, Millbank, London, S.W.1, a British Company, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to glass laminates.

More particularly, the invention relates to glass laminates having modified transparency, as compared with the glass itself, to provide useful or decorative effects.

In the term "glass" we include, as well as silicate glasses for which the term is conventionally used, transparent or translucent, substantially rigid sheet of plastics materials such as, for example, polymethyl methacrylate, polyvinyl chloride, polystyrene or polycarbonate resin.

Tinted glass, including that used for anti-glare windows and vehicle windscreens, may be made by incorporating colours or particles into the glass itself during manufacture. But this is an expensive process, and for most purposes tinted glass has been made by lamination, suitable additives being incorporated into the interlayer. However, this method has the disadvantage that the product does not give a high degree of protection from infra-red radiation.

In accordance with the present invention, a glass laminate comprises two sheets of glass (as hereinbefore defined) with a sheet of plastics film sandwiched between them, the plastics film being partially transparent and being bonded to both sheets of glass by a substantially transparent copolymer of ethylene having a melting point below that of the film. By "partially transparent" we mean that only parts of the film are transparent.

The invention also provides a method of making such a glass laminate that comprises forming a sandwich of a sheet of the partially transparent plastics film between two layers of glass (as hereinbefore defined), with a layer of the substantially transparent copolymer of ethylene having a melting point below that of the film between each sheet of glass and the adjacent film surface, heating the sandwich to a temperature above the melting point of the copolymer of ethylene but below that of the film, and thereafter holding the layers of the sandwich in contact with each other until the copolymer of ethylene has cooled to a solid state.

The film may provide colour or decoration to the laminate by having its surface printed, or by having a dye or particulate material incorporated into the plastics material from which it is made. In accordance with a particularly useful form of the invention, the plastics film is lightly metallized on at least one of its surfaces, so that, while remaining transparent in parts the laminate has anti-glare or anti-dazzle properties.

Biaxially oriented, heat set polyethylene terephthalate film is a preferred material for use in making the partially transparent films used in this invention since it has high clarity, high strength and dimensional stability, and melting point considerably higher than those of the transparent ethylene copolymers used for bonding to laminated safety glass. The thickness of the partially transparent film is not critical; generally it need not exceed 0.5 mm. Other materials

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that may be used include oriented polyamides.

The film may first be given a surface treatment to improve its bonding properties, such as, for example, by subjecting it to high voltage electric stress accompanied by corona discharge, particularly when it is to be printed or metallized.

Particularly preferred copolymers of ethylene useful in bonding the glass laminates of this invention are described in U.K. Specification No. 1,166,443. These are copolymers of ethylene with one or more hydroxy or epoxy aliphatic or cycloadiphatic monoesters of acrylic acid or methacrylic acid in which the aliphatic or cycloaliphatic radical consists only of carbon, hydrogen and oxygen and contains not more than six carbon atoms, and with from 0% to 55%, by weight of the copolymer, of a further comonomer having one ethylenic double bond which is an ester of acrylic or methacrylic acid or a vinyl ester, the copolymer containing, by weight, at least 35% of ethylene units and:

(a) from 2.0% to 8.5% of free hydroxy groups; or
(b) from 0.3% to 3.0% of epoxy oxygen; or
(c) an equivalently effective amount with respect to the percentages stated in (a) and (b), of both free hydroxy groups and epoxy oxygen.

Other copolymers of ethylene that may be used, include copolymers of ethylene with acrylic acid, and with a vinyl ester, an acrylate or a methacrylate as a third comonomer or copolymers of ethylene with methacrylic acid and with an acrylate or methacrylate as a third comonomer.

The layers of transparent copolymer of ethylene may be formed by coating the copolymer upon both surfaces of the plastics film layer either from solution or as a melt. Alternatively a 5-fold sandwich may be formed in which a film of the transparent copolymer of ethylene is sandwiched between opposed surfaces of the glass and the higher melting plastics film. When such a 5-fold sandwich is used in the method of the invention, a somewhat higher temperature or time of heating will generally be required than when the transparent copolymer of ethylene material has previously been coated on to the film.

In a widely used process for the production of safety glass, and suitable for making the laminates of this invention, the sheets of glass with the partially transparent film interlayer interposed between them are assembled in a suitable jig, and the assembly is placed in a flexible bag of plastics or rubber. The bag is then thoroughly evacuated, and the assembly is placed in an oven, or, preferably, in an air or oil autoclave, and heated to the desired bonding temperature. This is generally above 110°C, preferably between 110° and 170°C and the heating period is generally 15 to 30 miuntes. However higher temperatures and different time cycles may be used. The pressure of the atmosphere, when an oven is used, or the pressure applied in the autoclave, provides for uniform contact and bonding between the glass sheets and the interlayer. The assembly is then cooled or allowed to cool before its removal from the bag.

To obtain maximum transparency the laminate is preferably cooled as rapidly as is possible without risk of causing breakage of the glass. Alternatively, it is more convenient to allow the laminate to cool slowly, full transparency may be restored by reheating the laminate to a temperature of about 120°C and then rapidly cooling it. In either case, the laminates may be cooled by plunging them into liquid cooling medium, usually water, suitably at about 40°C. Cooling may alternatively be accomplished by means of blasts of cold air.

The laminates may also be made by applying heat and pressure in a mechanical or hydraulic press, with heating and cooling applied by heat transfer through the platens. The bag and autoclave method is generally much preferred for making laminates of high quality, particularly for making curved laminates.

The glass surfaces to be bonded may if desired be treated with an adhesion promoter before the sandwich is assembled. For example, silanes of the type described in United Kingdom Specification No. 1,095,700 may be used for this purpose.

The invention is of particular value when applied to the modification of transparent safety glass, especially that of the type used for windscreens for motor cars and other vehicles, of the type comprising two sheets of silicate glass, which may be curved or flat, with a layer of transparent bonding medium sandwiched between them. It is also useful when applied to the modification of a safety glass of the type comprising either a sheet or sheets of silicate glass bonded to a substantially rigid sheet of a transparent or translucent plastics material, or of the type comprising two or more substantially rigid sheets of transparent or translucent plastics material bonded together by the bonding medium. Such laminates are often used, for example, for bandit-resistant and bullet-resistant glass windows for shops, show cases, banks, vehicles and the like. The plastics material used to make the substantially rigid sheets is preferably an acrylic plastics material (especially polymethyl methacrylate) or a rigid polyvinyl chloride, or a polycarbonate resin. The modified safety-

glass laminates are especially useful when they are provided with anti-glare properties, by the use of a lightly metallized film. Such laminates give good protection from infrared radiation. The laminates in general may alternatively contain a tinted or printed film, such as one bearing a decorative pattern or advertising matter, when they may be used, for example, as panels for doors, partitioning walls or show cases.

Our invention is illustrated but in no way limited by the following Example.

EXAMPLE

A copolymer of ethylene with 12% of methacrylic acid and 14% of vinyl acetate by weight, the remainder being ethylene, was dissolved in toluene (30% solution at 70°C). This was coated on to each side of a layer of 0.003″ (0.075 mm) thick biaxially oriented polyethylene terephthalate film, lightly metallized on both sides so as to be partly transparent. The film was then laid between two layers of float glass, 0.125″ (3.06 mm) thick, with the tinned side of the glass towards the interlayer.

The sandwich was placed in a plasticised polyvinyl chloride bag, sealed and evacuated by means of a vacuum pump; it was then placed in an oven at 150°C, where it remained for 40 minutes and was then cooled.

The product had excellent optical and anti-glare properties.

WHAT WE CLAIM IS:—

1. A glass laminate comprising two sheets of glass (as hereinbefore defined) with a sheet of plastics film sandwiched between them, the plastics film being partially transparent and being bonded to both sheets of glass by a substantially transparent copolymer of ethylene having a melting point below that of the film.

2. A transparent glass laminate as claimed in Claim 1 in which the plastics film is lightly metallized on at least one of its surfaces to provide anti-dazzle properties.

3. A glass laminate as claimed in Claim 1 or Claim 2 in which the plastics film is a biaxially oriented, heat set film of polyethylene terephthalate.

4. A glass laminate as claimed in any one of the preceding claims in which the substantially transparent copolymer of ethylene is a copolymer of ethylene with one or more hydroxy or epoxy aliphatic or cycloaliphatic monoesters of acrylic acid or methacrylic acid in which the aliphatic or cycloadiphatic radical consists only of carbon, hydrogen and oxygen and contains not more than six carbon atoms, and with from 0% to 55%, by weight of the copolymer, of a further comonomer having one ethylenic double bond which is an ester of acrylic or methacrylic acid or a vinyl ester, said copolymer containing, by weight, at least 35% of ethylene units and:

(a) from 2.0% to 8.5% of free hydroxy groups; or
(b) from 0.3% to 3.0% of epoxy oxygen; or
(c) an equivalently effective amount with respect to the percentages stated in (a) and (b), of both free hydroxy groups and epoxy oxygen.

5. A glass laminate as claimed in any one of Claims 1 to 3 in which the substantially transparent copolymer of ethylene is a copolymer of ethylene with acrylic acid and with a vinyl ester, an acrylate or a methacrylate as a third comonmer.

6. A glass laminate as claimed in any one of Claims 1 to 3 in which the substantially transparent copolymer of ethylene is a copolymer of ethylene with methacrylic acid and with an acrylate or methacrylate as a third comonomer.

7. A glass laminate as claimed in any one of the preceding claims in which both sheets of glass are of silicate glass.

8. A glass laminate as claimed in any one of Claims 1 to 6 in which at least one of the glass sheets is a transparent or translucent, substantially rigid sheet of plastics material.

9. A glass laminate as claimed in Claim 1 and substantially as hereinbefore described in the Example.

10. A method of making a glass laminate as claimed in any one of Claims 1 to 9 that comprises forming a sandwich of a sheet of the partly transparent plastics film between two layers of glass (as hereinbefore defined), with a layer of the substantially transparent copolymer of ethylene having a melting point below that of the film between each sheet of glass and the adjacent film surface, heating the sandwich to a temperature above the melting point of the copolymer of ethylene but below that of the film, and thereafter holding the layers of the sandwich in contact with each other until the copolymer of ethylene has cooled to a solid state.

11. A method of making a glass laminate substantially as hereinbefore described with reference to the Example.

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